

AD 660212

ENGINEERING REPORT O. 8581

CONTINUOUS-WAVE STIMULATED
RAMAN EMISSION
FINAL REPORT

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PERKIN ELMER
OPTICAL GROUP NORWALK, CONNECTICUT

ENGINEERING REPORT NO. 8581

CONTINUOUS-WAVE STIMULATED
RAMAN EMISSION
FINAL REPORT

DATE: 30 JUNE 19

PREPARED FOR: CONTRACT NO. NoNr 4847(00)

ARPA ORDER NO. 306

PROJECT CODE NO. 4730

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SECTION I

INTRODUCTION

The goal of this program has been the elucidation of discrepancies between gains for stimulated Raman emission calculated from Q-switch laser experiments and those calculated from spontaneous Raman scattering cross-sections and lines widths. In principle, all of the features of stimulated Raman emission should be calculable from the latter two quantities^{1,2}. In practice, the observed gains for stimulated Raman emission were often found to be some ten times as large as those predicted from spontaneous Raman data³. The observed thresholds for stimulated Raman emission were often found to be correspondingly too low⁴. It was suggested, by Townes and coworkers⁵, that perhaps the anomalies were due to a beam-trapping phenomenon. If the intense electric fields in a Q-switched laser beam increased the refractive index in the medium, the beam would be trapped by total internal reflection. This would shrink the entire beam down to a filament by a cumulative process.

Within the scope of the present contract, two lines of approach were followed to test these ideas. One was an extensive theoretical⁶ and limited experimental⁷ study to explore beam trapping. The other was an attempt to measure the gain for stimulated Raman emission, at very low fields, with a continuous-wave argon laser. Our theoretical study explored electrostriction as a mechanism for refractive index change. The conclusion of the study was that electrostriction was probably important only in solids. We attempted to detect trapping of the entire laser beam in water, benzene, trichloroethylene, tetrachloroethylene, toluene, and 2-propanol using focused and unfocused beams.

Our photographs and measurements of beam diameter showed no evidence of entire beam trapping in these liquids. For later use, we measured the gain constant for stimulated Raman emission in the 1004 cm^{-1} line of toluene⁷.

The experiment for measuring low field stimulated Raman gains was the stimulated anti-Stokes experiment outlined in an earlier report⁸. Briefly, the idea was to cross, at the matching angle, the 4880 \AA and 5145 \AA argon laser lines in a liquid with a strong Raman shift at the wavenumber difference between these two lines (1057 cm^{-1}). Stimulated anti-Stokes emission and spontaneous anti-Stokes emission arising from the 4880 \AA line would be expected to appear at 4642 \AA . The spontaneous emission would be discriminated against by chopping the 5145 \AA line only and using synchronous detection.

Before the anti-Stokes experiment could be tried, a number of preliminary studies had to be made. An intensive literature search⁹ was made to locate compounds with a strong Raman emission near 1057 cm^{-1} . After preliminary screening, the spontaneous Raman spectra of the most promising compounds were obtained. The most promising of these were tested for stimulated Raman emission with a Q-switched laser¹⁰. In no case was stimulated Raman emission observed near 1057 cm^{-1} , but several of the substituted aromatics (in particular α - chloronaphthalene and 1,3 dibromobenzene) were found to give strong stimulated Raman emission at other frequencies. On the basis of the strength and narrow breadth of its 1057 cm^{-1} line and the ease with which it produced stimulated Raman emission at 1368 cm^{-1} , α - chloronaphthalene was chosen as the sample material for the experiment. Its dispersion was carefully measured on a large goniometer and matching angles calculated.

An optical train was developed for detaching the 4880 \AA and 5145 \AA argon lines from the other argon lines, crossing them at adjustable angles in

sample and bringing any anti-Stokes emission to the spectrometer. A device* was designed to suppress the "lens effect" due to heating of the samples by the laser beam.

Using the spontaneous Raman data and the gain constant measured in stimulated Raman emission in toluene, we estimated the yield of anti-Stokes radiation expected in our experiment. This estimate was well within the range of sensitivity of our experiment.

On trying the experiment, we found that Rayleigh scatter was so strong, that stray light in our monochromator effectively drowned the weak expected stimulated Raman emission anti-Stokes radiation. We were able to show, however (Section II of this report), that had an anomalously large gain been present, a signal would have appeared above noise.

At about this point in time, reports began to appear in the literature which seemed to explain the anomalous observed stimulated Raman emission as indeed being due to beam trapping.**

It was observed that beam trapping tends to occur in filaments, rather than over the whole beam¹². (This is not true in CS_2 , where whole-beam trapping has been observed¹³.) Although the subject is still being debated, it is probable that beam-trapping is indeed responsible for the observed anomalous gains⁴. The quadratic Kerr effect¹⁴, rather than electrostriction, apparently causes the refractive index changes.

In view of this work, and in view of our own demonstration that anomalous gain is absent in the low field case, it did not seem in the best interests of the contracting agency to pursue the actual measurement of the low-field gain. While such an experiment would be perfectly feasible, the knowledge that could be gained would not, at this point be worth the considerable expenditures of time and money required.

*Patent applied for.

**A review of the work to date is given by Minck, Terhune and Wang.¹¹

The demonstration that anomalous gains were not present in our experiment is given in Section II, following.

SECTION II

RESULTS

The final optical train and detection systems used are shown in Figure 1. The optical train shown in the 1 November 1965 - 31 January 1966 Quarterly Report 8 was modified by addition of a lens system to focus the output onto a pinhole and a lens to image the pinhole of the slit of a Jarrell-Ash 1/2 meter monochromator (Figure 1). The monochromator was set to the proper wavelength by viewing the spontaneous anti-Stokes emission (see below). The matching angle could then be scanned by moving the mirror mounted on the micrometer screw.

Use of this system guaranteed that the stimulated anti-Stokes radiation would get through the optics and into the spectrometer. The output from an EMI 6256SA phototube was fed through a PAR HR-8 lock-in amplifier into a Sanborn 8803A recorder.

Measurements were carried out using the 1057 cm^{-1} Raman line of α - chloronaphthalene.

By chopping the 4880 \AA beam instead of the 5145 \AA beam, it was possible to detect the spontaneous anti-Stokes line at -1057 cm^{-1} with a signal-to-noise ratio of about 5:1 when a 25 mm thick sample cell was used. With a 2 mm thick sample cell, nothing could be detected. This enables us at least to put some upper limits on the amount of stimulated anti-Stokes radiation that could have been detected with our system.

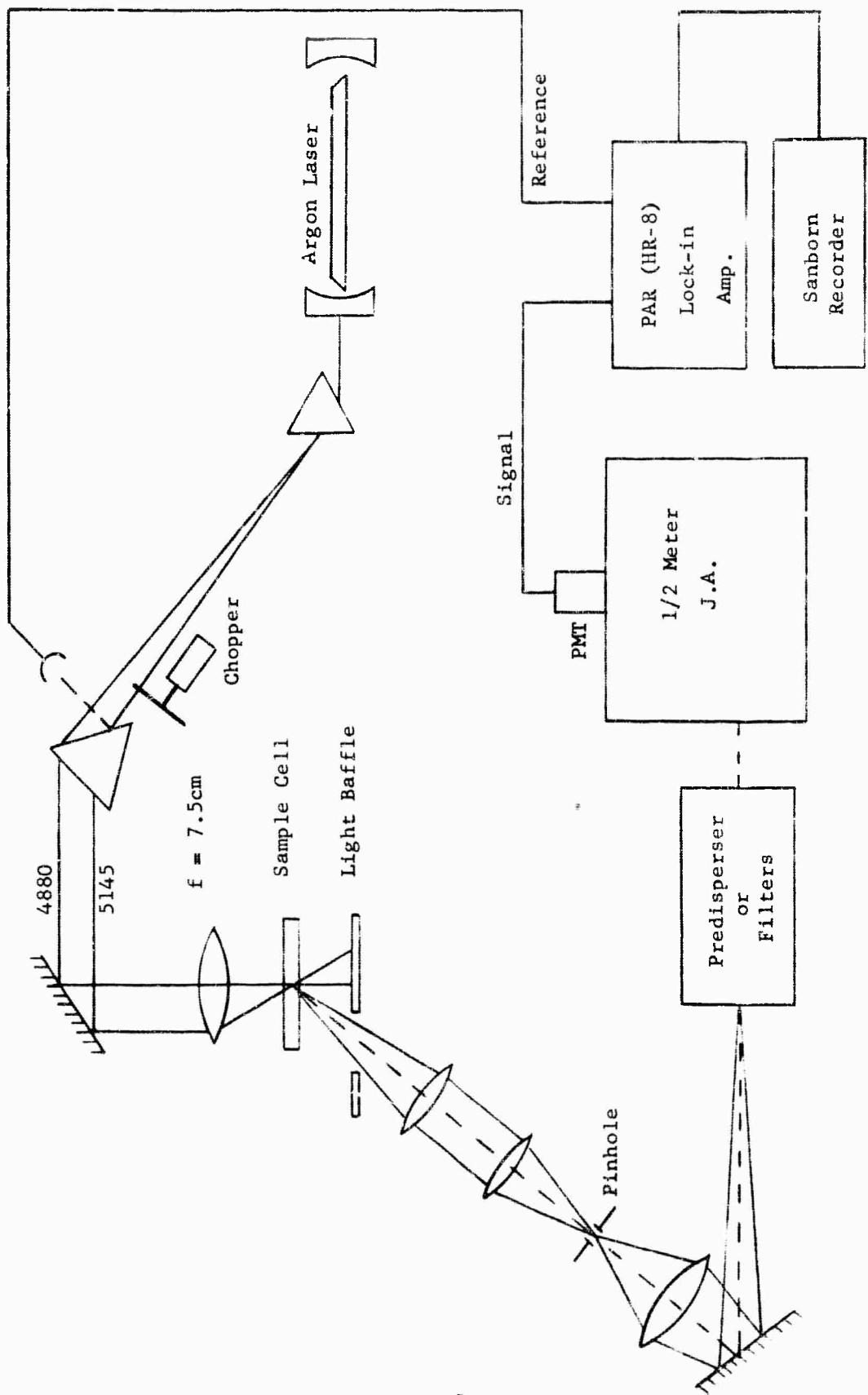


Figure 1. Optical Train for Stimulated Anti-Stokes Experiment

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With light of 4880 \AA and a lens of 7.3 cm focal length, the Fresnel volume is 10^{-5} cm^3 . Since α - chloronaphthalene has a molecular mass of 163 and a density of 1.63 gm cm^{-3} , the Fresnel volume contains 6×10^{16} molecules. The scattering cross-section for the 1057 cm^{-1} line should be about 10^{-29} cm^2 per molecule⁸. With 0.5 watt in the 4880 \AA line, the flux density into the Fresnel cross-section is about $10^4 \text{ watts cm}^{-2}$. The total power scattered into the 1057 cm^{-1} Stokes line from the Fresnel volume is $6 \times 10^{16} \times 10^{-29} \times 10^4 = 6 \times 10^{-9}$ watts.

The aperture stop of the spontaneous scattering was the light baffle after the sample cell. This had a hole 1 cm in diameter placed 20 cm from the sample cell. The subtended solid angle was, then, $3.14 \times (0.5)^2 / 400 = 0.002$ steradian. The scattering from the spontaneous Stokes at 1057 cm^{-1} into the spectrometer would have been $6 \times 10^{-9} \times 0.002/4 = 10^{-12}$ watts. At 300°K , for the 1057 cm^{-1} line, the scattering from the Fresnel volume into the spontaneous anti-Stokes is $(7 \times 10^{-3}) \times 10^{-12} = 7 \times 10^{-15}$ watts. (The Fresnel length was about 0.2 cm, the length of the short sample cell.)

The spontaneous anti-Stokes scattering just calculated may be compared with the expected value of 2.8×10^{-14} watts for stimulated anti-Stokes emission calculated in the 1 November 1965 - 31 January 1966 Quarterly Report. The latter figure was based on an estimated gain for stimulated Stokes emission of 0.005 cm^{-1} at a field of $2.5 \times 10^7 \text{ watts cm}^{-2}$.

We were unable to see either spontaneous (chopping 4880 \AA) or stimulated (chopping 5145 \AA) anti-Stokes emission above noise, using the 0.2 cm sample cell. Our system was therefore not capable of seeing 10^{-14} watts above noise.

We were able to see the spontaneous anti-Stokes emission using a 25 mm long sample cell. Due to uncertainties in the geometry, and the fact that the illuminating flux density was not constant outside the Fresnel region, the output from this cell cannot be estimated exactly. We know for certain that it was less than 10 times as great as that from the 2 mm cell, so that a power output in the stimulated anti-Stokes emission of 7×10^{-14} watts or more could have been detected with a signal-to-noise ratio of 5:1. Taking into account the uncertainties in our estimates, it seems that a stimulated anti-Stokes signal of 2.8×10^{-14} watts should have been just at the edge of detectability. Therefore, our gain for stimulated emission was less than or equal to $4.25 \times 10^{-6} \text{ cm}^{-1}$ and we were not in the anomalous gain region. (At the $2.5 \times 10^7 \text{ watts cm}^{-2}$ of the Q-switched laser experiment, the corresponding gain would be 0.005 cm^{-1} .)

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SECTION III

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Unclassified

Security Classification

DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)

Perkin-Elmer
Electro-Optical Division
Norwalk, Connecticut, 06852

2a. REPORT SECURITY CLASSIFICATION

2b. GROUP

3. REPORT TITLE

CONTINUOUS-WAVE STIMULATED RAMAN EMISSION

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Final Report

5. AUTHOR(S) (Last name, first name initial)

Tobin, Marvin C.
Barrett, Joseph J.

6. REPORT DATE

30 June 1967

7a. TOTAL NO. OF PAGES

8

7b. NO. OF REFS

14

8a. CONTRACT OR GRANT NO.

NoNr 4847(00)

9a. ORIGINATOR'S REPORT NUMBER(S)

b. PROJECT NO.
4730

Perkin-Elmer Engineering
Report No. 8581

c. TASK

d.

9b. OTHER REPORT NO(S) (Any other numbers that may be
assigned this report)

10. AVAILABILITY LIMITATION NOTICES

11. SUPPLEMENTARY NOTES

12. SPONSORING MILITARY ACTIVITY

Advanced Research Products Agency
Washington, D.C.

13. ABSTRACT

The goal of this program has been the elucidation of discrepancies between gains for stimulated Raman emission calculated from Q-switch laser experiments and those calculated from spontaneous Raman scattering cross-sections and line widths.

Within the scope of the contract, two lines of approach were followed to test theories and ideas previously advanced by other workers in the field. One approach was a study to explore beam trapping by extensive theoretical and limited experimental means. The second was an attempt to measure the gain for stimulated Raman emission, at very low fields, with a continuous-wave argon laser.

The conclusions were that beam-trapping was responsible for the observed anomalous gains and that no anomalous gain was present at low fields.

Unclassified**Security Classification**

14.

KEY WORDS**Q-Switched laser****Raman Emission**

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